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ABSTRACT:

An impact polypropylene composition comprising: (i) isotactic polypropylene; and (ii) a copolymer of ethylene and an alpha-olefin comonomer having 3 to 8 carbon atoms, said copolymer (a) having a density of about 0.87 to about 0.90 gram per cubic centimeter and a crystallinity in the range of about 20 to about 35 percent by weight based on the weight of the copolymer and (b) being present in the composition in the range of about 20 to about 50 percent by weight based on the weight of the polypropylene.

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⑤ Impact polypropylene.

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EP-A- 0 052 557
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GB-A- 1 346 234
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Description

This invention relates to an improved version of impact polypropylene, particularly a product molded therefrom.

6 Impact polypropylene is widely used in such applications as appliances, automobiles, furniture, and luggage. It is generally a blend of isotactic polypropylene with ethylene/propylene or ethylene/propylene/diene rubbers or a similar combination prepared in situ. The rubber modifier overcomes the low temperature brittleness and the lack of impact resistance of the isotactic polypropylene. However, when the polypropylene is modified with an amorphous rubber, it experiences a severe stress whitening or 10 blush on impact, e.g., when an object made of this impact polypropylene is struck, a white discoloration appears. This is obviously undesirable as it limits the applications of the impact polypropylene.

15 GB-A-2 060 658 and 2 083 278 describe compositions for the production of cold drawn film. Said compositions comprise an ethylene- α -olefin copolymer elastomer having a density of not more than 0.91 g/cm³ and at least one polymer selected from crystalline polypropylene and polybutene-1 as essential components.

16 EP-A-168 129 is directed to a polypropylene composition having superior clarity and good low-temperature impact resistance and comprising crystalline polypropylene and ethylene-propylene random copolymer containing at least 70 wt % of ethylene.

An object of this invention is to provide a molded product of polypropylene, which exhibits much 20 reduced blushing together with high impact resistance.

Other objects and advantages will become apparent hereinafter.

According to the present invention, the above object is met by a product molded from a composition comprising the following components:

25 (i) isotactic polypropylene; and
(ii) a copolymer of ethylene and 1-butene, said copolymer (a) having a density of about 0.87 to about 0.90 gram per cubic centimeter and a crystallinity in the range of about 20 to about 35 percent by weight based on the weight of the copolymer and (b) being present in the composition in a range of about 20 to about 50 percent by weight based on the weight of the polypropylene.

30 Isotactic polypropylene homopolymer can be prepared by the process described in US-A-4,304,891. The homopolymer preferably has a melt flow in the range of about 1 to about 20 and xylene solubles in the range of about 2 to about 6.

35 The ethylene/1-butene copolymer can be prepared by the processes described in EP-A-120 501 and 120 503. The density of the copolymer is in the range of about 0.87 to about 0.90 gram per cubic centimeter. The portion of the copolymer attributed to the comonomer (1-butene) is in the range of up to about 40 mole percent based on the total number of moles in the copolymer and is preferably in the range of about 7 to about 30 mole percent. The balance of the copolymer is based on ethylene. The portion of the copolymer based on 1-butene is in the range of about 15 to about 60 percent by weight based on the weight of the copolymer, and is preferably in the range of about 20 to about 45 percent by weight. These ethylene/1-butene copolymers are considered to be soft polymers because they are semi-crystalline. The 40 crystallinity is in the range of about 20 to about 35 percent by weight. The copolymer has a melt index in the range of about 0.1 to about 10 grams per 10 minutes and preferably in the range of about 0.3 to about 1.0 gram per 10 minutes. Melt index is determined by ASTM D-1238, Condition E. It is measured at 180 °C. The isotactic polypropylene and the ethylene/1-butene copolymers are, of course, not reactive with one another.

45 Subject composition can be prepared either by physical blending or in situ incorporation of the copolymer in a polypropylene matrix. The proportion of ethylene/1-butene copolymer can be in the range of about 20 percent by weight to about 50 percent by weight based on the weight of the isotactic polypropylene.

50 A typical blending technique is described as follows: isotactic polypropylene is mixed (dry) with the ethylene/1-butene copolymer and the dry mixture is extruded at about 200 °C and pelleted. Conventional stabilizers for the isotactic polypropylene and the ethylene/1-butene copolymers are used.

55 A typical in-situ method is carried out in the following manner: Isotactic polypropylene homopolymer is prepared in one reactor and is transferred to another reactor (or reactors) where the ethylene/1-butene copolymer is produced in the presence of the polypropylene homopolymer. The in-situ mixture is taken out of the reactor(s), stabilized, and extruded at about 200 °C and pelleted.

Various conventional additives can be added in conventional amounts to subject compositions such as antioxidants, ultraviolet absorbers, antistatic agents, pigments, dyes, fillers including carbon black, slip agents, fire retardants, stabilizers and smoke inhibitors.

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The invention is illustrated by the following examples:

Examples 1 to 6

5 A polypropylene homopolymer having a melt flow of 4.0 and xylene solubles of 3.5 is blended with an ethylene/propylene copolymer or an ethylene/1-butene copolymer wherein the portion of the copolymer based on comonomer, propylene or 1-butene, is present in amounts of 25 and 15 percent by weight, respectively, based on the weight of the copolymer. The copolymer is present in the blend in an amount of 15 to 50 percent by weight based on the weight of the polypropylene. The blend is extruded and pelletized 10 with the following stabilizers: 0.125 weight percent calcium stearate and 0.125 weight percent antioxidant. The pellets are injection molded into 3.175 mm (125 mil) specimens for evaluation.

The Table sets forth the variables, i.e., the weight percent of the copolymer based on the weight of the composition; the density of the copolymer in gram per cubic centimeter; and the crystallinity in percent by weight, and the results.

15 The tests used to obtain the results are as follows:

1. The Gardner Impact (-30°C) test is carried out according to ASTM D-3029. This test involves dropping a weight from a defined height onto an injection molded disk, 3.175 mm (125 mil) thick, which has been preconditioned at -30°C. The results are given in cm-kg (inch-lbs).
2. The 1% SFM (Secant Flexural Modulus) test is carried out according to ASTM D-780. The results are given in MPa (psi).
- 20 3. The blushing [4.53 kg (10 lbs)] test is carried out as follows. An injection molded disk is used. The apparatus is the same as for the Gardner Impact test except that the falling weight (a steel bar) is 4.53 kg (10 pounds). The steel bar is dropped on the disk. The disk is then aged at room temperature for 24 hours. Stress-whitening or blushing appears on the disk in the form a circle. The extent of stress-whitening is defined by the diameter of the circle measured in fractions of a cm (in).
- 25 4. Crystallinity is measured by Differential Scanning Colorimeter (DSC) using a Dupont 990 analyzer with a pressure DSC cell.

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Table

Example	Comonomer	% Co-Polymer	Density (g/cc)	Crystallinity (% by wt.)	Gardner Impact cm-kg (inch-lbs)	1% SFM MPa (psi)	Blushing at 4.53kg cm (in)
1*	propylene	30	0.86	<2	>388 (>320)	760 (110,000)	1.80 (0.63)
2*	propylene	15	0.87	20	23 (20)	1,100 (180,000)	1.27 (0.5)
3	1-butene	30	0.87	20	>345 (>300)	880 (125,000)	1.02 (0.4)
4	1-butene	50	0.87	20	>345 (>320)	580 (88,000)	0.0 (0.0)
5	1-butene	30	0.80	35	201 (175)	830 (120,000)	0.68 (0.26)
6*	1-butene	30	0.82	45	23 (20)	900 (130,000)	<0.25 (<0.1)
7*	1-butene	30	0.88	89	<12 (<10)	1,280 (188,000)	0.0 (0.0)

* = Comparative

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Melt flow is determined in accordance with ASTM-1638.

Xylene solubles are defined as the fraction that stays in solution after the polypropylene sample is dissolved in hot xylene and the solution is allowed to cool to 23°C.

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Claims

1. Product molded from a composition comprising:
 - (i) Isotactic polypropylene; and
 - (ii) a copolymer of ethylene and 1-butene, said copolymer (a) having a density of about 0.87 to about 0.90 gram per cubic centimeter and a crystallinity in the range of about 20 to about 35 percent by weight based on the weight of the copolymer and (b) being present in the composition in the range of about 20 to about 50 percent by weight based on the weight of the polypropylene.
- 15 2. The molded product defined in claim 1, which exhibits a blush of at most about 1.27 cm (0.5 inch) per 4.53 kg (10 pounds).
3. The molded product defined in one or both of the claims 1 to 2 wherein the portion of the copolymer based on 1-butene is present in the range of about 15 to about 60 percent by weight based on the weight of the copolymer.
- 20 4. The molded product defined in claim 3 wherein the portion of the copolymer based on 1-butene is present in the range of about 20 to about 45 percent by weight based on the weight of the copolymer.

25 Revendications

1. Produit moulé à partir d'une composition comprenant :
 - (i) un polypropylène isotactique ; et
 - (ii) un copolymère d'éthylène et de 1-butène, ledit copolymère (a) ayant une masse volumique d'environ 0,87 à environ 0,90 g/cm³ et une cristallinité comprise dans l'intervalle d'environ 20 à environ 35% en poids, sur la base du poids du copolymère, et (b) étant présent dans la composition en une quantité comprise dans l'intervalle d'environ 20 à environ 50% en poids, sur la base du poids du polypropylène.
- 35 2. Produit moulé suivant la revendication 1, qui présente un voile d'eau plus environ 1,27 cm (0,5 in) pour 4,53 kg (10 lbs).
3. Produit moulé suivant l'une, ou bien l'une et l'autre, des revendications 1 et 2, dans lequel la portion du copolymère à base de 1-butène est présente en une quantité comprise dans l'intervalle d'environ 15 à environ 60% en poids, sur la base du poids du copolymère.
- 40 4. Produit moulé suivant la revendication 3, dans lequel la portion du copolymère à base de 1-butène est présente en une quantité comprise dans l'intervalle d'environ 20 à environ 45% en poids, sur la base du poids du copolymère.

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Patentansprüche

1. Produkt, geformt aus einer Zusammensetzung, die umfaßt:
 - (i) isotaktisches Polypropylen und
 - (ii) ein Copolymer von Ethylen und 1-Buten, das (a) eine Dichte von etwa 0,87 bis etwa 0,90 g/cm³ und eine Kristallinität im Bereich von etwa 20 bis etwa 35 Gew.-%, bezogen auf das Gewicht des Copolymers, hat und (b) in der Zusammensetzung in einer Menge von etwa 20 bis etwa 50 Gew.-%, bezogen auf das Gewicht des Polypropylens, vorliegt.
- 55 2. Formprodukt gemäß Anspruch 1, das ein Anlaufen von höchstens etwa 1,27 cm (0,5 inch) pro 4,53 kg (10 pounds) zeigt.
3. Formprodukt gemäß einem oder beiden der Ansprüche 1 bis 2, worin der Anteil des auf 1-Buten

basierenden Copolymers im Bereich von etwa 15 bis etwa 60 Gew.-%, bezogen auf das Gewicht des Copolymers, vorliegt.

4. Formprodukt gemäß Anspruch 3, wovon der Anteil des auf 1-Buten basierenden Copolymers im Bereich von etwa 20 bis etwa 45 Gew.-%, bezogen auf das Gewicht des Copolymers, vorliegt.

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